

FORM PTO-1390
REV. 2/01T

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

05788.0199

U.S. APPLICATION NO.
(If known, see 37CFR1.5)

10/019447

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/EP00/06203

July 3, 2000

July 1, 1999


TITLE OF INVENTION**WAVEGUIDE SYSTEMS OR STRUCTURES OR PARTS THEREOF, CONTAINING POLYCYANATE
COPOLYMERS PREPARED FROM POLYFUNCTIONAL CYANATES AND FLUORINATED MONOCYANATES****APPLICANT(S) FOR DO/EO/US****Monika BAUER, Jörg BAUER, Christian DREYER, Norbert KEIL and Crispin ZAWADZKI**

Applicant(s) herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c)(2)).
 - a. ☒ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed with the United States Receiving Office (RO/US).
- ☐ An English language translation of the International Application as filed (35 U.S.C. 371 (c)(2)).
 - a. ☐ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154 (d)(4).
- ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)).
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
- ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
- ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
- ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

ems 11 to 20 below concern document(s) or information included:

- ☒ Information Disclosure Statement under 37 CFR 1.97 and 1.98
- ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
- ☒ A FIRST preliminary amendment.
- ☐ A SECOND or SUBSEQUENT preliminary amendment.
- ☐ A Substitute specification.
- ☐ A change of power of attorney and/or address letter.
- ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825.
- ☐ A second copy of the published international application under 35 U.S.C. 154 (d)(4).
- ☐ A second copy of the English language translation of the international application 35 U.S.C. 154 (d)(4).
- ☒ Other items or information:
 - a. ☒ Copy of cover page of International Publication No. WO 01/02464 A1
 - b. ☐ Copy of Notification of Missing Requirements.
 - c. ☐

U.S. APPLICATION NO. (If known, see 37CFR 1.5) 10/019447		INTERNATIONAL APPLICATION NO. PCT/EP00/06203		ATTORNEY'S DOCKET NUMBER: 05788.0199	
21. <input checked="" type="checkbox"/> The following fees are submitted:				CALCULATIONS PTO USE ONLY	
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):					
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1040.00					
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890.00					
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00					
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00					
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00					
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). <input type="checkbox"/> 20 <input type="checkbox"/> 30				\$	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total Claims	33	- 20 =	13	x \$18.00	\$234.00
Independent Claims	1	- 3 =		x \$84.00	\$
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+ \$280.00	\$280.00
TOTAL OF THE ABOVE CALCULATIONS =				\$1404.00	
<input checked="" type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$	
SUBTOTAL =				\$1404.00	
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest priority date (37 CFR 1.492(f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30				\$	
TOTAL NATIONAL FEE =				\$1404.00	
Fee for recording the enclosed assignment (37 CFR 1.21 (h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property.				\$	
TOTAL FEES ENCLOSED =				\$1404.00	
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<p><input checked="" type="checkbox"/> A check in the amount of \$ <u>1404.00</u> to cover the above fees is enclosed.</p> <p><input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p><input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>06-0916</u>. A duplicate copy of this sheet is enclosed.</p> <p><input type="checkbox"/> Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.</p>					
<p>TE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) it be filed and granted to restore the application to pending status.</p>					
<p>TO ALL CORRESPONDENCE TO: egan, Henderson, Farabow, Garrett & Dunner, L.L.P. 1111 I Street, N.W. Washington, D.C. 20005-3315</p>					
<p>3D: December 31, 2001</p>				<p> SIGNATURE Ernest F. Chapman / 25,961 NAME/REGISTRATION NO.</p>	

PATENT
Customer No. 22,852
Attorney Docket No. 05788.0199

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:)
)
Monika BAUER et al.) Group Art Unit: Not Yet Assigned
)
Serial No.: Unassigned) Examiner: Not Yet Assigned
)
PCT Filed: July 3, 2000)
)
U.S. National Stage Filed: December 31, 2001)
)
For: **WAVEGUIDE SYSTEMS OR
STRUCTURES OR PARTS THEREOF,
CONTAINING POLYCYANATE
COPOLYMERS PREPARED FROM
POLYFUNCTIONAL CYANATES AND
FLUORINATED MONOCYANATES**

**National Phase Filing of International
Application No. PCT/EP00/06203**

Box PCT
Assistant Commissioner for Patents
Washington, DC 20231

Sir:

PRELIMINARY AMENDMENT

Prior to the examination of the above application, please amend this application
as follows:

IN THE SPECIFICATION:

Please amend the specification as follows:

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Page 1, after the title, please insert the following new headings and subheading and new paragraph:

--Cross Reference to Related Applications

This application is a national stage filing of International Application No. PCT/EP00/06203, filed July 3, 2000, which published in the English language and claims priority of European Patent Application No. 99112596.4, filed on July 1, 1999.

BACKGROUND OF THE INVENTION

Field of the Invention--.

Page 1, before line 37, add the following new subheading:

--Description of the Related Art--.

Page 3, before line 16, add the following new section heading:

--SUMMARY OF THE INVENTION--.

Page 3, before line 31, add the following new section heading:

--DETAILED DESCRIPTION OF THE INVENTION--.

IN THE CLAIMS:

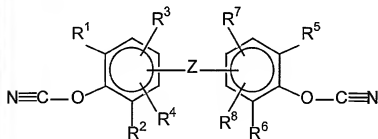
Please cancel now pending claims 1-16 without prejudice or disclaimer and substitute new claims 17-29 therefor as follows:

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WHAT IS CLAIMED IS:

17. (New) An optical waveguide system or a structure or part thereof, comprising a resin composed of at least one polycyanate copolymer, obtainable by copolymerization of at least one difunctional cyanate of formula II:



(II)

wherein R¹ to R⁴ and R⁵ to R⁸ are independently from each other hydrogen, optionally substituted C₁-C₁₀ alkyl, C₃-C₈-cycloalkyl, C₁-C₁₀-alkoxy, halogen, phenyl or phenoxy, the alkyl or aryl groups being unfluorinated, partly fluorinated or fully fluorinated, Z is a chemical bond, SO₂, CF₂CH₂, CHF, CH(CH₃), isopropylene, hexafluoroisopropylene, n- or iso-C₁-C₁₀ alkylene, O, NR⁹, N=N, CH=CH, C(O)O, CH=N, CH=N-N=CH, alkyl oxyalkylene having 1 to 8 carbon atoms, S, Si(CH₃)₂, and R⁹ is hydrogen or C₁-C₁₀ alkyl

with at least one monocyanate of the following formula I:



(I)

wherein R is a straight or branched non-aromatic hydrocarbon radical or a non-aromatic hydrocarbon radical comprising a cyclic structure, the radical having the formula C(R')₂-CFR'₂ wherein each R' is, independently from the other, hydrogen or fluorine or an

optionally substituted alkyl or alkenyl group, and each of R" may independently be defined as R' or may have an aryl structure.

18. (New) An optical waveguide system or a structure or part thereof according to claim 17, wherein the substituted alkyl or alkenyl group of R' is fluorinated.

19. (New) An optical waveguide system or a structure or part thereof according to claim 17, wherein the polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanate of formula I and at least one dicyanate having formula III:



wherein R¹⁰ is a non-aromatic hydrocarbon group carrying at least one fluorine atom.

20. (New) An optical waveguide system or a structure or part thereof according to claim 19, wherein R¹⁰ of formula III is a partly or fully fluorinated alkylene group having 1 to 15 carbon atoms.

21. (New) An optical waveguide system or a structure or part thereof according to claim 20, wherein the partly or fully fluorinated alkylene group has 3 to 12 carbon atoms.

22. (New) An optical waveguide system or a structure or part thereof according to claim 19, wherein the polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanate of formula I, optionally at least one dicyanate having formula III, and a monocyanate of formula IV

US 2005/0148481 A1

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temperature of from 100°C to 300°C and a refractive index of about 1.35 to about 1.60 at 1.55µm.

28. (New) An optical waveguide system or a structure or part thereof according to any one of claims 17 to 27, wherein the system, structure or part thereof is an optical fibre, a waveguide, a buffer layer, a cladding or a support for any of said structures.

29. (New) An optical waveguide system comprising a waveguide consisting of a resin as defined in any one of claims 17 to 27, and a buffer or cladding consisting of a resin as defined in any one of claims 17 to 27, but different from the resin of the waveguide, wherein the resin of the waveguide has a greater refractive index than that of the buffer or cladding.

REMARKS

In this Preliminary Amendment, the claims have been amended to conform them to U.S. practice and to eliminate improper multiple claim dependency.

Claims 17-29 are pending in this application. No new matter has been added.

If there is any fee due in connection with the filing of this Preliminary Amendment, please charge the fee to our Deposit Account No. 06-0916.

Respectfully submitted,

FINNEGAN, HENDERSON, FARABOW,
GARRETT & DUNNER, L.L.P.

Dated: December 31, 2001

By: 

Ernest F. Chapman
Reg. No. 25,961

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(19) World Intellectual Property Organization
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PCT

(10) International Publication Number
WO 01/02464 A1(51) International Patent Classification: C08G 73/06,
G02B 1/04(74) Common Representative: PIRELLI CAVI E SISTEMI
S.P.A.; Viale Sarca, 222, I-20126 Milano (IT).

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1 July 1999 (01.07.1999) EP(71) Applicant (*for all designated States except US*): PIRELLI CAVI E SISTEMI S.P.A. [IT/IT]; Viale Sarca, 222, I-20126 Milano (IT).(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

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Published:

- With international search report.
- Before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: WAVEGUIDE SYSTEMS OR STRUCTURES OR PARTS THEREOF, CONTAINING POLYCYANATE COPOLYMERS PREPARED FROM POLYFUNCTIONAL CYANATES AND FLUORINATED MONOCYANATES

(57) Abstract: The present invention is directed to wave guide systems or structures or parts thereof, characterized in that they consist of or comprise a resin composed of at least one polycyanate copolymer, obtainable by copolymerization of at least one specific difunctional cyanate with at least one monocyanate of the formula $N=C-O-R$, wherein R is a straight or branched non-aromatic hydrocarbon radical or a non-aromatic hydrocarbon radical comprising a cyclic structure, the radical having the formula $C(R')_2-CFR''_2$ wherein each R' is, independently from the other, hydrogen or fluorine or an optionally substituted, preferably fluorinated alkyl or alkenyl group, and each of R'' may independently be defined as R' or may have an aryl structure. The at least one difunctional cyanate is selected from aromatic dicyanates having two aryl rings, connected with each other by a group Z wherein Z is a chemical bond, SO_2 , CF_2 , CH_2 , CHF , $CH(CH_3)$, isopropylene, hexafluoroisopropylene, n- or iso- C_1-C_{10} alkylene, O, NR^2 , $N=N$, $CH=CH$, $C(O)O$, $CH=N$, $CH=N-N=CH$, alkyl oxyalkylene having 1 to 8 carbon atoms, S, $Si(CH_3)_2$, and R^2 is hydrogen or C_1-C_{10} alkyl. The polycyanate copolymer may further comprise an aromatic monocyanate and/or one or more of brominated cyanates. The polycyanate copolymers are advantageously selected for the preparation of optical waveguide systems or structures or parts thereof because they have low optical losses at 1.3 and 1.55 μm .

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Waveguide systems or structures or parts thereof, containing polycyanate copolymers prepared from polyfunctional cyanates and fluorinated monocyanates

- 5 The present invention is directed to optical elements in the field of waveguide systems or waveguide structures, e.g. arrayed wave guide components, prepared by copolymerization of specific polyfunctional cyanates and fluorinated monocyanates, as well as to the use of said copolymers for the preparation of said
- 10 structures.

Organic polymers are increasingly interesting materials in the optical or microoptical field, in integrated optics or in microsystem techniques. In these fields, they may be used in

15 optical instruments and apparatuses or parts thereof as well as in special optics as lenses, prisms, for fixation of optical systems, as support material for optical layers and as translucent coating materials for mirrors or lenses. Organic polymers may be used in optical fibres and for the preparation

20 of waveguide structures. Their technical handling is relatively easy, and their density is lower in comparison to glass.

Specifically, if such plastics or organic polymers are to be used as a waveguide, a variety of requirements are to be met.

- 25 The refractive index of the material should be variable in a range as broad as possible and should be adaptable to that of specific substrates. If used in the optical communication engineering, low absorptions of the materials are required at 1.3 and 1.55 μ m. The loss due to attenuation caused by volume
- 30 defects (non-homogenities, microbubbles, microfissures) should be minimized. Besides specific technological requirements, e.g. preparation of layers and structurability, specific provisions for the use of organic polymers as waveguide structures in integrated optics are the thermal and thermo-mechanical
- 35 stability, adapted extension coefficients and long term stability of optical properties.

Until now, polymethacrylates and polycarbonates have been mainly used for optical purposes. Both classes of polymers have an

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excellent light transmittance, but their thermal and thermo-mechanical stability is not sufficient due to their chemical structure. Thus, polymethacrylates and polycarbonates cannot practically be used at temperatures exceeding 130°C which is due to their relatively low glass transition temperatures. In addition, both types of polymers are linear, un-crosslinked polymers. This has the adverse effect that they are partly solubilized in case multilayer-systems are prepared via the application of dissolved components, e.g. by spin-coating each layer. Consequently, the layer structures as obtained are not sufficiently delimited and neat which, however, is an essential for the preparation of waveguide structures.

There are other high performance polymers which have glass transition temperatures of more than 180°C. Examples are polyarylethersulfones, polyarylsulfones, polyaryletherketones, polyimides and polyetherimides, the processing of which, however, is more difficult than that of polymethacrylates and polycarbonates. Another disadvantage of these systems is the relatively high optical loss at wave lengths of 1.3 and 1.55µm, relevant in communication engineering.

Polyperfluorocyclobutanes (PFCB) are a relatively new class of high performance polymers. Upon thermal curing they yield insoluble cross-linked polymers which are characterized by high thermal stability. Waveguide layers prepared from PFCB show very low optical losses of 0.2 dB/cm at 1550 nm.

Also, polycyanurates have been used for the preparation of optical components. US patents 5 208 892 and 5 165 959 describe the preparation of polycyanate resins made of a single monomer (either fluorinated or non-fluorinated). German Offenlegungsschrift DE 44 35 992 A1 describes optical elements prepared from polycyanurate resins. The resins are made by polymerization of dicyanate or polycyanate compounds, optionally in mixture with di- or polyphenols or di- or polyglycidyl compounds. Like polyperfluorocyclobutanes, polycyanurates yield insoluble cross-linked polymers upon thermal curing, and these

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polymers are as well characterized by high thermal stability. They are specifically useful due to their excellent adhesional force on a variety of substrates, for example silicon, silica or a variety of organic polymers. Refractive index and glass transition temperature of the cured cross-linked polymers may be varied in broad ranges, due to the easy availability of a great number of di- and mono-functional cyanate monomers which may be copolymerized with each other. Polycyanurates of the kind mentioned above are partly commercially available. Completely cured polycyanurates known in the art which consequently are stable for long terms may have optical losses of about 0.2 dB/cm at 1.3 μ m. However, the optical losses are not less than 0.5dB/cm at 1.55 μ m which is important in communication engineering technologies.

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The present invention provides copolymers, obtainable by copolymerization of at least one monocyanate, derived from a partly or fully fluorinated alcohol ("fluorinated monocyanate"), and at least one specific difunctional organic cyanate. It has been found that such copolymers are specifically valuable in the preparation of optical waveguide systems or structures thereof having low optical losses at 1.3 and at 1.55 μ m.

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30
Throughout the invention, "partly fluorinated" means that at least one fluorine atom is present in the molecule. "Fully fluorinated" means that hydrogen atoms are completely substituted by fluorine atoms. The whole molecules, or single organic radicals or groups (e.g. methyl, methylene, alkyl, aryl groups), respectively, may be fully fluorinated.

As fluorinated monocyanate, one, two, three or even more monocyanates of formula I may be used

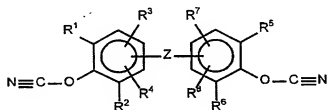


(I)

35
wherein R is $\text{C}(\text{R}')_2\text{-CFR}''_2$, wherein each R' is, independently from the other, hydrogen or fluorine or an optionally substituted, preferably fluorinated alkyl or alkenyl group having preferably

1 to 13, more preferably 3 to 11 carbon atoms. Each of R" may independently be defined as R'. Further, R" may have an aryl structure. Preferably, R is a straight, branched, or cyclic non-aromatic hydrocarbon radical or an non-aromatic hydrocarbon radical comprising a cyclic structure. Preferably, the non-aromatic hydrocarbon radical has 1 to 15, more preferably 3 to 12 carbon atoms. It is to note that each of the carbon atoms of R may carry 1, 2 or, if it is a terminal carbon atom, 3 fluorine atoms. Fully fluorinated carbon atoms (-CF₃, -CF₂-) are preferred. Further, it is preferred that one or both of R' are hydrogen and/or one of R" is fluorine or a partly or fully fluorinated alkyl and the other is a partly or, more preferable, fully fluorinated alkyl which may be straight, branched or cyclic. Specific examples for the cyanates of formula (I) are -CH₂-CF₂-CF₃, -CH₂-CF₂-CF₂-CF₃, -CH₂-C(CF₃)₂F, -CH₂-CF₂-CF₂-CF₃.

For the preparation of the said copolymer, one, two, three or even more difunctional organic cyanates may be used. The expression "difunctional" means that two NCO groups are present in the molecule. The NCO groups are bound to organic radicals via the oxygen atom. The difunctional cyanate may be, but is not necessarily, partly or fully fluorinated. The organic structure of the difunctional cyanate or cyanates is selected under difunctional cyanates of formula II:



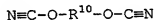
(II)

wherein R¹ to R⁴ and R⁵ to R⁸ are independently from each other hydrogen, optionally substituted C₁-C₁₀ alkyl, C₃-C₈-cycloalkyl, C₁-C₁₀-alkoxy, halogen, phenyl or phenoxy, the alkyl or aryl groups being unfluorinated, partly fluorinated or fully fluorinated, Z is a chemical bond, SO₂, CF₂ CH₂, CHF, CH(CH₃), isopropylene, hexafluoroisopropylene, n- or iso-C₁-C₁₀ alkylene

which may be partly or fully fluorinated, O, NR⁹ with R⁹ being hydrogen or C₁-C₁₀ alkyl, N=N, CH=CH, C(O)O, CH=N, CH=N-N=CH, alkoxyalkylene having 1 to 8 carbon atoms which is optionally partly or fully fluorinated, S, or Si(CH₃)₂. Examples are

- 2,2'-bis(4-cyanato-phenyl)propane,
- 2,2'-bis(4-cyanato-phenyl)hexafluoropropane,
- biphenylene-4,4'-dicyanate,
- 2,3,5,6,2',3',5',6'-octafluorobiphenylene-4,4'-dicyanate.

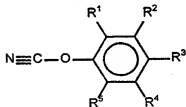
In one embodiment of the invention, dicyanates according to formula III:



(III)

wherein R¹⁰ is an organic non-aromatic hydrocarbon group carrying at least 1 fluorine atom are copolymerized into the polycyanate copolymer useful in the present invention. In formula (III), R¹⁰ is preferably an alkylene group, more preferably having 3 to 12 carbon atoms. Each of the carbon atoms may carry 0, 1 or 2 or, in the case of a terminal group, 3 fluorine atoms. The carbon chain may be straight or branched or may be cyclic or may contain a cyclic part. Further, it may contain one or more C=C double bonds. In one embodiment, R¹⁰ is fully fluorinated. Examples are -CH₂-CF₂-CF₂-CH₂- or -CH₂-CF₂-CF₂-CF₂-CF₂-CH₂.

In another embodiment of the present invention, at least one additional monocyanate having formula IV:



(IV)

wherein R¹ to R⁵ are as previously defined for formula II, is copolymerized in addition to the starting cyanates as defined above (with or without (a) cyanate(s) of formula III) in order to obtain the polycyanate copolymer. Examples for compounds of formula IV are phenylcyanate and perfluorophenylcyanate.

Specifically, the refractive index and the glass transition temperature may be influenced by this additive as desired.

The polycyanate copolymers according to the invention may be obtained by mixing at least one of the monocyanates of formula I, optionally in addition to at least one of formula IV, and at least one difunctional organic cyanate of formula II, optionally in addition to at least one of formula III. The ratio of monocyanates to dicyanates may be freely chosen, provided that at least 1% by mol, preferably at least to 5% by mol, more preferably at least 10% by mol of monocyanate of structure I is present per mol of monomers to be polymerized. Preferably, the monofunctional cyanates of formulas I and IV are present in a molar amount of not more than 75% related to the total amount of moles of monomers present in the mixture to be copolymerized.

The starting monocyanate and dicyanate compounds as described above are preferably warmed up after mixing. The temperature may be chosen as required; a range of about 120°C to 170°C is preferred. Preferably, the reaction is performed in the absence of oxygen, e.g. in a sealed and preferably (under an inert gas atmosphere). The mixture is allowed to react until a liquid or viscous prepolymer (resin) is obtained. This prepolymer or resin is soluble in useful solvents, preferably in solvents having high polarity, e.g. ethylethoxyacetate or chlorobenzene. In general, the prepolymer is processed in a respective solution, e.g. by spin-coating of a solution containing 25 to 65% by weight of the prepolymer, more preferably about 50% by weight of the prepolymer. The prepolymer solution may be applied to a suitable substrate, consisting of e.g. silicon, quartz or an organic polymer. After being brought into the desired shape (e.g. a layer of desired thickness) it is cured (e.g. at temperatures in the range of 200° to 260°C) in order to provide the desired network between the cyanate groups.

If an optical wave guide system comprising a variety of different layers of the present polycyanate copolymers shall be

prepared, each different layer is applied and is cured, e.g. thermically cured, before the next layer is applied.

It shall be clear that the term "resin" is independent of the condition of the polymer, e.g. whether it is in a prepolymerized condition or is partly or completely cured.

The polycyanate copolymers according to the present invention have a glass transition temperature in the range of 100° to 300°C, and their refractive index at 1.55μm may be controlled in the desired range, specifically of from 1.35 to 1.60. Specifically, the more fluorinated monomers are used, or the more fluorine parts per weight are present in the mixture, related to the weight of the mixture to be polymerized, the lower is the refractive index of the polycyanate copolymer obtained.

On the other hand, use of brominated derivatives of the cyanate monomers as defined above will raise the refractive index of the copolymer obtained. Thus, monocyanate compounds of e.g. formula IV wherein at least one of R¹, R², R³, R⁴ or R⁵ is substituted by bromine, may be advantageously added to the mixture. In general, the more bromine is included in the polymer, the higher is the refractive index obtained. Accordingly, any of the cyanates of formulas I to IV as defined above (with the proviso that those of formula I may be free of fluorine) carrying one or more bromine atoms may be selected. However, brominated monocyanates are preferably used, either alone or in mixture with brominated polycyanates.

The polycyanate copolymers according to the present invention are used for the preparation of optical wave guide systems or parts thereof. For example, they may be used for the preparation of waveguides and waveguide structures. For such structures, use of at least two different polycyanate copolymers is preferred, wherein a polycyanate copolymer having a lower refractive index may be used for buffer and/or cladding while a polycyanate copolymer differing from the first one and having a greater

refractive index may be used as the optical waveguide. At least one of these polycyanate copolymers should have been obtained according to the present invention. The selection will be easily made by a skilled person who is able to control the refractive index via the teachings given in this application. The layers show excellent adhesion to each other and to the substrate. Waveguide structures as described above may be prepared by known methods, e.g. RIE (Reactive Ion Etching).

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The invention is now further illustrated by way of examples.

Example 1

5 12.9 g of a substituted dicyanate of Bisphenol A (compound II wherein R^1-R^4 is H, R^5-R^8 is H, Z is hexafluoroisopropyl) and 3.7 g of a partly fluorinated monocyanate (compound I wherein R is $CH_2-CF_2-CF_2-CF_3$) are heated to $160^\circ C$ in a sealed vessel for a time of about four hours. The reaction is terminated before gelling
10 starts, and a clear, pale yellow prepolymer is obtained which is viscous at $160^\circ C$ and is solid at room temperature. The prepolymer is brought into solution by mixing it with 50 % by weight of ethylethoxyacetate (EEA). Spin-coating of this
15 solution onto a substrate made of silicon wafer yields a layer which may be cured at $240^\circ C$ for one hour in a drying oven. The product has a refractive index of 1.4776 at $1.55 \mu m$.

Example 2

20 12.9 g of a substituted dicyanate of Bisphenol A (compound II wherein R^1-R^4 is H, R^5-R^8 is H, Z is hexafluoroisopropyl), 3.7 g of a partly fluorinated monocyanate (compound I wherein R is $CH_2-CF_2-CF_2-CF_3$), and 1.3 g of a monocyanate (compound IV wherein R^1 , R^2 , R^4 , R^5 are hydrogen and R^3 is bromine) are heated to $160^\circ C$ in
25 a sealed vessel for a time of about four hours. The reaction is terminated before gelling starts, and a clear, pale yellow prepolymer is obtained which is viscous at $160^\circ C$ and is solid at room temperature. The prepolymer is brought into solution by
30 mixing it with 50 % by weight of EEA. Spin-coating of this solution onto a substrate made of silicon wafer yields a layer which may be cured at $240^\circ C$ for one hour in a drying oven. The product has a refractive index of 1.4870 at $1.55 \mu m$.

Example 3

9.7 g of dicyanate of Bisphenol A (compound II wherein R^1-R^4 is H, R^5-R^8 is H, Z is isopropyl) and 2.5 g of a fully fluorinated monocyanoate (compound I wherein R is C-(CF₃)₂) are heated to 140°C in a sealed vessel for a time of about four hours. The reaction is terminated before gelling starts, and a clear, pale yellow prepolymer is obtained which is viscous at 140°C and is solid at room temperature. The prepolymer is brought into solution by mixing it with 50 % by weight of EEA. Spin-coating of this solution onto a substrate made of silicon wafer yields a layer which may be cured at 240°C for one hour in a drying oven. The product has a refractive index of 1.5596 at 1.55 μ m.

Example 4

9.7 g of a substituted dicyanate of Bisphenol A (compound II wherein R^1-R^4 is H, R^5-R^8 is H, Z is hexafluoroisopropyl), 10.3 g of a partly fluorinated dicyanate (compound III wherein R^{10} is CH₂-CF₂-CF₂-CF₂-CF₂-CH₂) and 1.1 g of a partly fluorinated monocyanoate (compound I wherein R is CH₂-CF₂-CF₂-CF₃) are heated to 140°C in a sealed vessel for a time of about four hours. The reaction is terminated before gelling starts, and a clear, pale yellow prepolymer is obtained which is viscous at 140°C and is solid at room temperature. The prepolymer is brought into solution by mixing it with 50 % by weight of EEA. Spin-coating of this solution onto a substrate made of silicon wafer yields a layer which is cured at 240°C for one hour in a drying oven. The product has a refractive index of 1.3689 at 1.55 μ m.

Example 5

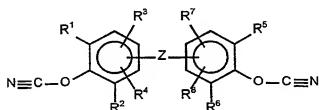
A 50 weight-% solution of the prepolymer of example 1 in EEA is spin-coated onto a silicon wafer, yielding a layer of about 8 μm thickness. Curing is performed at 240°C in a drying oven for one hour. Onto this layer, a 50 weight-% solution of the prepolymer of example 2 in EEA is spin-coated, again yielding a layer of about 8 μm thickness. Also, this layer is cured at 240°C in the drying oven for about 1 hour. According to known methods, an aluminum layer of about 100 nm is sputtered onto the said second prepolymer layer followed by its structurization by way of photolithography and chemical etching. Subsequently, the waveguides are structured by aid of oxygen RIE techniques (typical rate 100 nm/min using pure oxygen), and the etching mask is removed by treatment in a chemical etching bath. Then, the upper cladding layer is applied by spin-coating a prepolymer solution of example 1 followed by curing at 240°C for 1 hour. Using near field technique a difference of 0.0094 of the refractive index between the waveguide and its surrounding is measured. Cut-back measurements of light intensities of waveguides of different lenght yielded a loss of 0.35 dB/cm at 1.55 μm .

* * *

Claims:

1. Optical waveguide system or a structure or part thereof, comprising a resin composed of at least one polycyanate copolymer, obtainable by copolymerization of at least

one difunctional cyanate of formula II:



(II)

wherein R^1 to R^4 and R^5 to R^8 are independently from each other hydrogen, optionally substituted C_1 - C_{10} alkyl, C_3 - C_8 -cycloalkyl, C_1 - C_{10} -alkoxy, halogen, phenyl or phenoxy, the alkyl or aryl groups being unfluorinated, partly fluorinated or fully fluorinated, Z is a chemical bond, SO_2 , CF_2 CH_2 , CHF, $CH(CH_3)$, isopropylene, hexafluoroisopropylene, n- or iso- C_1 - C_{10} alkylene, O, NR^9 , $N=N$, $CH=CH$, $C(O)O$, $CH=N$, $CH=N-CH$, alkyl oxyalkylene having 1 to 8 carbon atoms, S, $Si(CH_3)_2$, and R^9 is hydrogen or C_1 - C_{10} alkyl

with at least one monocyanate of the following formula I:



(I)

wherein R is a straight or branched non-aromatic hydrocarbon radical or a non-aromatic hydrocarbon radical comprising a cyclic structure, the radical having the formula $C(R')_2-CFR''_2$ wherein each R' is, independently from the other, hydrogen or fluorine or an optionally substituted, preferably fluorinated

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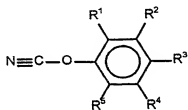
alkyl or alkenyl group, and each of R" may independently be defined as R' or may have an aryl structure.

2. Optical waveguide system or a structure or part thereof according to claim 1, characterized in that the said polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanate of formula I and at least one dicyanate having formula III:



wherein R¹⁰ is a non-aromatic hydrocarbon group carrying at least one fluorine atom.

3. Optical waveguide system or a structure or part thereof according to claim 2, characterized in that R¹⁰ of formula III is a partly or fully fluorinated alkylene group having 1 to 15, more preferably 3 to 12 carbon atoms.
4. Optical waveguides system or a structure or part thereof according to any of the preceding claims, characterized in that the said polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanate of formula I, optionally at least one dicyanate having formula III, and a monocyanate of formula IV

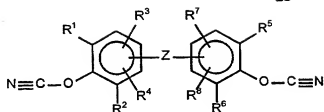


(IV)

wherein R¹ to R⁵ are defined as in formula II.

5. Optical waveguide system or a structure or part thereof according to any of the preceding claims, characterized in that the said polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanat of formula I, and at least one brominated monocyanate of formulas I to III, preferably of formula I, as defined above with the proviso that the monocyanates of formula I may be free of fluorine.
6. Optical waveguide system or a structure or part thereof according to any of the preceding claims, characterized in that the monocyanate of formula I as defined in claim 1 is used in an amount of at least 10%, preferably of at least 20% by mol per mol of the polycyanate copolymer.
7. Optical waveguide or a structure or part thereof according to any of the preceding claims, characterized in that the polycyanate copolymer has a glass transition temperature of from 100°C to 300°C and/or has a refractive index of about 1.35 to about 1.60 at 1.55µm.
8. Optical waveguide system or a structure or part thereof according to any of the preceding claims, characterized in that it is an optical fibre, a waveguide, a buffer layer, a cladding or a support for any of the said structures.
9. Optical waveguide system comprising a waveguide consisting of a resin composed as defined in any of claims 1 to 7, and a buffer and/or cladding consisting of a resin composed of a resin as defined in any of claims 1 to 7, but different from that of the waveguide, wherein the resin of the waveguide has a greater refractive index than that of the buffer and/or cladding.
10. Use of a polycyanate copolymer, obtainable by copolymerization of at least one polyfunctional cyanate selected from a difunctional cyanate of formula II:

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(II)

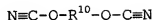
wherein R^1 to R^4 and R^5 to R^8 are independently from each other hydrogen, optionally substituted C_1 - C_{10} alkyl, C_3 - C_8 -cycloalkyl, C_1 - C_{10} -alkoxy, halogen, phenyl or phenoxy, the alkyl or aryl groups being unfluorinated, partly fluorinated or fully fluorinated, Z is a chemical bond, SO_2 , CF_2 CH_2 , CHF , $CH(CH_3)$, isopropylene, hexafluoroisopropylene, n- or iso- C_1 - C_{10} alkylene, O, NR^9 , $N=N$, $CH=CH$, $C(O)O$, $CH=N$, $CH=N-N=CH$, alkyl oxyalkylene having 1 to 8 carbon atoms, S, $Si(CH_3)_2$, and R^9 is hydrogen or C_1 - C_{10} alkyl

with at least one monocyanate of the following formula I:



wherein R is a straight or branched non-aromatic hydrocarbon radical or a non-aromatic hydrocarbon radical comprising a cyclic structure, the radical having the formula $C(R')_2-CFR''_2$ wherein each R' is, independently from the other, hydrogen or fluorine or an optionally substituted, preferably fluorinated alkyl or alkenyl group, and each of R'' may independently be defined as R' or may have an aryl structure, as a material in optical waveguide systems or structures or parts thereof, preferably of optical fibres, waveguides, buffers, claddings, or supports for such structures.

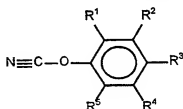
11. Use of a polycyanate copolymer according to claim 10, characterized in that the said polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanat of formula I and at least one dicyanate having formula III:



(III)

wherein R^{10} is a non-aromatic hydrocarbon group carrying at least one fluorine atom, preferably a partly or fully fluorinated alkylene group having 1 to 15, more preferably 3 to 12 carbon atoms.

12. Use of a polycyanate copolymer according to claim 10 or 11, characterized in that the said polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanate of formula I, optionally at least one dicyanate having formula III, and a monocyanate of formula IV



(IV)

wherein R^1 to R^5 are defined as in formula II.

13. Use of a polycyanate copolymer according to any of claims 10 to 12, characterized in that the said polycyanate copolymer is obtainable by copolymerization of at least one difunctional cyanate of formula II, at least one monofunctional cyanate of formula I, and at least one brominated monocyanate of formulas I to III, preferably of formula I, as defined above with the proviso that the monocyanates of formula I may be free of fluorine.

14. Use of a polycyanate copolymer according to any of claims 10 to 13, characterized in that the monocyanate of formula I as defined in claim 1 is used in an amount of at least 10%, preferably of at least 20% by mol per mol of the polycyanate copolymer.

15. Use of a polycyanate copolymer according to any of claims 10 to 14, characterized in that the said polycyanate copolymer has a glass transition temperature of from 100°C to 300°C and/or has a refractive index of about 1.35 to about 1.60 at 1.55µm.

16. Use of a polycyanate copolymer according to any of claims 10 to 15, wherein at least the materials used for the optical waveguide and for the buffer/and or cladding are those as defined in claim 10 and the material for the waveguide has a grater refractive index than that of the buffer and/or cladding.

* * *

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DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; I believe I am the original, first, and sole inventor (if only one name is listed below) or an original, first, and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**WAVEGUIDE SYSTEMS OR STRUCTURES OR PARTS THEREOF, CONTAINING POLYCYANATE COPOLYMERS
PREPARED FROM POLYFUNCTIONAL CYANATES AND FLUORINATED MONOCYANATES**

the specification of which

☐ is attached and/or

X was filed on December 31, 2001 as United States Application Serial No. 10/019,447 and was amended on December 31, 2001 or

X PCT International Application No. PCT/EP00/06203, filed on July 3, 2000.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate or § 365(a) of any PCT International application(s) designating at least one country other than the United States, listed below and have also identified below, any foreign application(s) for patent or inventor's certificate, or any PCT International application(s) having a filing date before that of the application(s) of which priority is claimed:

Country	Application Number	Date of Filing	Priority Claimed Under 35 U.S.C. 119
Europe	99112596.4	July 1, 1999	X YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below:

Application Number	Date of Filing

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s) or § 365(c) of any PCT International application(s) designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application(s) in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application(s) and the national or PCT International filing date of this application:

Application Number	Date of Filing	Status (Patented, Pending, Abandoned)

I hereby appoint the following attorney and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. **FINNegan, Henderson, Farabow, Garrett & Dunner, L.L.P., CUSTOMER NUMBER 22,852**, Douglas B. Henderson, Reg. No. 20,291; Ford F. Farabow, Jr., Reg. No. 20,630; Arthur S. Garrett, Reg. No. 20,338; Donald R. Dunner, Reg. No. 19,073; Brian G. Brunsvold, Reg. No. 22,593; Tipton D. Jennings, IV, Reg. No. 20,645; Jerry D. Voight, Reg. No. 23,020; Laurence R. Heffer, Reg. No. 20,827; Kenneth E. Payne, Reg. No. 23,098; Herbert H. Mintz, Reg. No. 26,691; C. Larry O'Rourke, Reg. No. 26,014; Albert J. Santorelli, Reg. No. 22,610; Michael C. Elmer, Reg. No. 25,857; Richard H. Smith, Reg. No. 20,609; Stephen L. Peterson, Reg. No. 26,325; John M. Romary, Reg. No. 26,331; Bruce C. Zottler, Reg. No. 27,680; Dennis P. O'Reilly, Reg. No. 27,932; Allen M. Sokal, Reg. No. 26,695; Robert D. Bajefsky, Reg. No. 25,387; Richard L. Stroup, Reg. No. 28,478; David W. Hill, Reg. No. 28,220; Thomas L. Irving, Reg. No. 28,619; Charles E. Lipsey, Reg. No. 28,165; Thomas W. Winland, Reg. No. 27,605; Basil J. Lewis, Reg. No. 28,818; Martin I. Fuchs, Reg. No. 28,508; E. Robert Yoches, Reg. No. 30,120; Barry W. Graham, Reg. No. 29,924; Susan Haberman Griffen, Reg. No. 30,907; Richard B. Racine, Reg. No. 30,415; Thomas H. Jenkins, Reg. No. 30,857; Robert E. Converse, Jr., Reg. No. 27,432; Lair X. Mullen, Jr., Reg. No. 20,348; Christopher P. Foley, Reg. No. 31,354; John C. Paul, Reg. No. 30,413; Roger D. Taylor, Reg. No. 28,992; David M. Kelly, Reg. No. 30,953; Kenneth J. Meyers, Reg. No. 25,146; Carol P. Elinaudi, Reg. No. 32,220; Walter Y. Boyd, Jr., Reg. No. 31,738; Steven M. Anzalone, Reg. No. 32,095; Jean B. Fordis, Reg. No. 32,984; Barbara C. McCurdy, Reg. No. 32,120; James K. Hammond, Reg. No. 31,964; Richard V. Burguljan, Reg. No. 31,744; J. Michael Jakes, Reg. No. 32,824; Thomas W. Banks, Reg. No. 32,719; Christopher P. Isaac, Reg. No. 32,616; Bryan C. Diner, Reg. No. 32,409; M. Paul Barker, Reg. No. 32,013; Andrew Chanho Sonu, Reg. No. 33,457; David S. Forman, Reg. No. 33,694; Vincent P.

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